Water Vapor Diffusion Through Soil as Affected by Temperature and Aggregate Size

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Abstract   Water vapor diffusion through the soil is an important part in the total water flux in the unsaturated zone of arid or semiarid regions and has several significant agricultural and engineering applications because soil moisture contents near the surface are relatively low. Water vapor diffusing through dry soil is absorbed for both long and short terms. Long-term absorption allows more water to enter than exit the soil, as reflected in the concentration gradient. Short-term absorption leads to an apparent reduction in the diffusion rate, as reflected in the diffusion coefficient. This investigation studied the effects of soil temperature and porosity on the isothermal diffusion of water vapor through soil. The diffusion model consisted of 25.4 cm × 8.9 cm × 20.3 cm Plexiglas box divided into two compartments by a partition holding a soil reservoir. Water vapor moved from a container suspended by a spring in one compartment, through the porous medium in the center of the model, to calcium chloride in a container suspended by a spring in the other compartment. The porous materials consisted of aggregates of varying size (2–2.8, 1–2, and 0.5–1 mm) of a Fayatte silty clay loam (a fine-silty, mixed mesic Typic Hapludalf). The flow rates of water vapor were measured at temperatures of 10, 20, 30, and 40°C. Warmer temperatures increased the rate of diffusion through dry soil while reduced the amount of water absorbed by that soil. Reducing porosity slowed the rate of diffusion and increased the amount of water absorbed. The dry soil in this study absorbed from 1/8 to 2/3 of the diffusing water. Maximum absorption rates occurred with the most compact soil samples at the highest temperature, though the maximum absorption as a percentage of the diffusing water was in the compact samples at the lowest temperature. The diffusivity equation $D/D_0 = [(S - 0.1)/0.9]^2$ fit the D/D_0 values obtained from these data if a coefficient of 1/3 or 1/3.5 is added to correct for the time delays caused by temporary sorption of the diffusing water vapor. The data, influenced by the interaction of water vapor

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and soil materials, represent a diffusion rate lower than the diffusion rate that would have resulted without this interaction.

Keywords   Diffusion coefficient · Absorption · Thermal gradient · Fick's law

1 Introduction

In arid and semiarid regions, water vapor movement is often an important part of the total water flux and energy balance in the vadose zone in many agricultural and engineering applications since soil moisture contents near the surface usually are very low (Milly 1984; Parlange et al. 1998).

In agricultural applications, water vapor diffusion is important since the actual contact area between liquid water and seeds is often very small such that seeds need imbibe water from vapor to germinate (Wuest et al. 1999).

In engineering applications, understanding surface energy, water vapor, and heat movement in the soil is critical for the performance evaluation of engineered surface covers for waste containment in landfills in arid and semiarid regions (Scanlon et al. 2005). Another engineering application requiring an assessment of coupled water vapor and heat transport involves performance of the potential of nuclear waste repositories. Generated heat may cause evaporation of soil water, and subsequent migration and condensation of water vapor in cooler areas (Spycher et al. 2003).

Water vapor moves through unsaturated soils in response to either thermal gradients or concentration gradients. Research has shown that increasing temperature increases the total quantity of water vapor diffusing through soil in response to concentration gradients. Even in dry soils, some moisture movement takes place in the vapor form and plays an important role in soil water regimes (Gurr et al. 1952; Jackson 1963; Onchukov et al. 1972).

Early pioneering studies, including Taylor and Cavazza (1954), Rollins et al. (1954), Kuzmak and Sereda (1957), and Matthes and Bowen (1963), have demonstrated that moisture movement in response to a thermal gradient through an unsaturated soil occurs mainly in the vapor phase. In fact, experimentally observed values for water vapor diffusion under thermal gradients are several times higher than those predicted by a simple diffusion equation (Philip and DeVries 1957).

Jackson (1963, 1964a,b) studied temperature and pressure effects on sorption diffusion coefficients. He concluded that vapor diffusion with the associated evaporation–condensation process is the predominant water transport mechanism in coarse-textured and dry soils. In fine-textured soils, liquid flow in thin water films and along particle and peds surfaces is appreciable at high water contents.

Water vapor movement in soil under isothermal conditions has also been measured (Onchukov et al. 1972). The results indicated that two processes take place in the initial period at a temperature of 20°C and a low initial water content of 6.5%; sorption of water vapor by the soil and vapor migration to the soil surface under the effect of the partial pressure gradient.

Accurate measurement of diffusion rates is facilitated by the use of gases that do not interact with the soil solids and liquids or other materials used in the experiment. Various gases have been used for this reason such as hydrogen (Currie 1984), freon (Sallam et al. 1984), and ethane (Pritchard 1985). These and several other researchers have measured the rate of diffusion of gases through soil and other porous media. The data are reported either

as diffusion rates or as diffusion coefficients. A number of these from some of the older data are summarized by Currie (1960).

Tokunaga et al. (1988) developed a transient, closed-tube method for measuring oxygen diffusivity in air-dry soil. Their results agreed well with values obtained by other methods as reported in the literature.

Various authors including Penman (1940), Taylor (1949), Currie (1960), Millington and Quirk (1960), Ayers et al. (1942), and Lai et al. (1976) have used either a linear equation such as $D/D_0 = a(S - b)$ or a curvilinear equation of the form $D/D_0 = KS^m$ to approximate their data (D = diffusion rate through the porous medium, D_0 = diffusion rate through the air, D/D_0 is the relative gas diffusivity (diffusion coefficient), S = open pore space (air-filled porosity of soil), and a, b, K, and m are constants whose values make the equations approximate the data). Troeh et al. (1982) compiled parameters for these diffusion equations to fit the results of several authors and proposed a new equation for such data. This equation takes the form $D/D_0 = [(S - u)/(1 - u)]^v$, where D, D_0, and S are defined as above and u and v are the fitting parameters. This equation is developed to allow for a fraction of the pore space equal to the u parameter being ineffective for diffusion (such as with isolated air pockets), and to provide the curved form of the exponential equation (controlled by the v parameter). It is the only one of the three types that necessarily results in $D = D_0$ when $S = 1$. The Troeh et al. (1982) model is considered useful for accurately fitting and evaluating soil measured data within the S interval where measured data are available but cannot at present be used to estimate gas diffusivity (Moldrup et al. 2005). Furthermore, Petersen et al. (1994) and Jin and Jury (1996) found that Troeh et al. (1982) model could describe experimental data better than any other model. They concluded that the Troeh et al. (1982) model is highly flexible, so that most experimental data can be satisfactory fitted, when both soil-specific parameters (u and v) are optimized simultaneously.

More recent D/D_0 models have begun to include parameters reflecting soil physical and hydraulic properties, such as total porosity (Millington and Quirk 1960; Moldrup et al. 2004; Fujikawa and Miyazaki 2005) and water retention (Moldrup et al. 1996, 2000), and some soil type-dependent models have become well established. These models provide a reasonable prediction of D/D_0 by using soil type-specific models. However, the comprehensive evaluation of soil physical properties for D/D_0 is still in progress (Fujikawa and Miyazaki 2005).

The objectives of this study were (1) to examine the effects of soil temperature and porosity on the isothermal diffusion of water vapor through soil in response to a concentration gradient, and (2) to fit the diffusivity model developed by Troeh et al. (1982) into diffusion coefficients resulted from this study.

2 Theory

The absorption of water vapor by the soil causes the amount of water vapor diffusion to vary with the position in the sample. This variation can be handled mathematically by adding a sink term, α, to the equation for Fick's second law of gas diffusion in differential form (Crank 1956) to obtain:

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2}\right) - \alpha \tag{1}$$

where C is the concentration of water vapor ($g\,cm^{-3}$), t is the time (s), D is the diffusion coefficient ($cm^2\,s^{-1}$), x is the distance of travel through the soil sample (cm), and α is water

absorption rate per unit volume of soil ($g \, cm^{-3} s^{-1}$). From its definition, α can be calculated as water absorbed (g)/(time (s) × soil volume (cm^3)). For a steady state condition, Eq. 1 can be set equal to zero and solved to give:

$$\frac{d^2C}{dx^2} = \frac{\alpha}{D} \tag{2}$$

Integration of Eq. 2 gives:

$$\frac{dC}{dx} = \alpha \frac{x}{D} + \text{constant} \tag{3}$$

The constant of integration can be evaluated by defining q_a as the rate of water absorption by the CaCl$_2$ ($g \, cm^{-2} s^{-1}$) and observing that $\frac{dC}{dx} = -\frac{q_a}{D}$ at the upper surface of the sample where $x = L$ (the sample thickness, cm). Thus

$$\frac{dC}{dx} = \alpha \frac{x}{D} - \alpha \frac{L}{D} - \frac{q_a}{D} \tag{4}$$

Rearranging Eq. 4, integrating, and evaluating the constant gives:

$$C = -\frac{1}{D} \left(\alpha L x - \frac{\alpha x^2}{2} + q_a x \right) + C_w \tag{5}$$

where C is the water vapor concentration anywhere within the soil sample, and C_w is the value of C at $x = 0$ (on the water side). Solving Eq. 5 for D gives:

$$D = \left(\alpha L x - \frac{\alpha x^2}{2} + q_a x \right) \Big/ (C_w - C) \tag{6}$$

Evaluating Eq. 6 for the exit side where $x = L$ and $C = C_e$

$$D = \left(q_a L + \frac{\alpha L^2}{2} \right) \Big/ (C_w - C_e) \tag{7}$$

Values of D are commonly converted to the dimensionless form D/D_0 and graphed accordingly (See Sect. 4). For this purpose, D/D_0 values for the diffusion rate of water vapor in the absence of any obstruction can be calculated from the expression $D_0 = 0.220(T/273)^{1.75}$ to be 0.2343, 0.2490, 0.2640, and 0.2795 $cm^2 s^{-1}$ for 10, 20, 30, and 40°C, respectively, where T is the absolute temperature (°K) (Handbook of Chemistry and Physics 2006).

The following assumptions were made when interpreting data: (i) water vapor flow is steady state (verified by measurements taken at five or more successive time intervals for each run), (ii) the water vapor absorption by the soil is linear across the samples, (iii) the water content is sufficiently low that liquid water flow was negligible, and (iv) Fick's law is valid throughout the flow medium. This last assumption requires some interpretation. Some of the water was absorbed within the samples, and it seems certain that much of the water vapor that passed through the samples was slowed by being adsorbed and desorbed along the way. Thus the diffusion rate determined is an apparent rate that includes this sorption time delay as well as the actual vapor diffusion time. The author speculates that this is what actually happens in natural soils and that this apparent diffusion rate is more meaningful for water vapor than the diffusion rate determined with a non-interactive gas-medium combination.

3 Materials and Methods

The diffusion laboratory model was a Plexiglas box with inside dimensions of 25.4 cm × 8.9 cm × 20.3 cm. It was divided into two parts by a partition holding a soil reservoir. Water vapor moved from a container suspended by a spring in one compartment, through the porous medium in the center of the model, to calcium chloride in a container suspended by a spring in the other compartment (Fig. 1). A controlled temperature was maintained by keeping the apparatus inside a Percival Chamber.

The porous materials used were various sizes of aggregates obtained by sieving a Fayette silty clay loam B horizon (a fine-silty, mixed mesic Typic Hapludalf). The aggregates sizes were 2.8–2 mm, 2–1 mm, and 1–0.5 mm. Table 1 shows some physical properties of the porous media used in this study. The samples used were air dry and contained 0.3–0.35% water at the beginning of each set of measurements and <5% water at the end of each set.

The porous-medium reservoir consisted of a ring of Plexiglas with an inside diameter of 5.08 cm and with a 200-mesh screen mounted below it. The screen was covered by a cloth mesh screen to support a layer of soil 0.64 cm thick. The sample reservoir was inserted into a hole in the partition in the middle of the Plexiglas box. A measured volume of tap water was placed in the pan to the right side of the sample, and the pan on the left side was filled with anhydrous granular calcium chloride drying agent. The Plexiglas box was sealed by applying petroleum jelly to the joining surfaces and attaching the cover with four screws.

The controlled-temperature chamber was set at the desired temperature, and the system was allowed to equilibrate for periods of 24 h for the 10 and 20°C trials and 12 h for the 30 and 40°C trials before any measurements were made. The brass springs were calibrated and their changes in length were measured to determine the weight of water evaporated on the one side and the amount of water absorbed by the calcium chloride on the other side. The
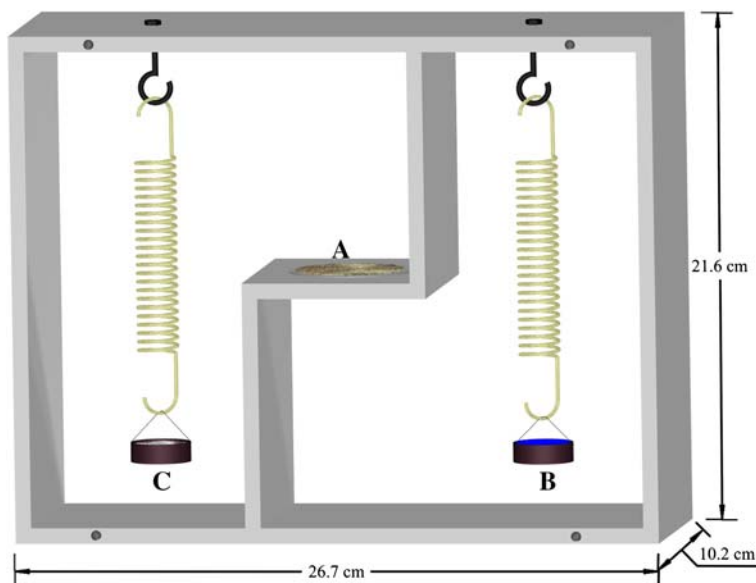

Fig. 1 The diffusion chamber was made of clear Plexiglas with soil in a reservoir (**A**) in the center. Water vapor moved from the pan (**B**) on the right through the soil to the calcium chloride in the container (**C**) on the left

Table 1 Selected physical properties of the Fayette silty clay loam B horizon soil material used in the diffusion experiment

Physical property	Size of aggregates			
	2–2.8 mm	1–2 mm	0.5–1 mm	0.5–1 mm (compacted)
Texture		Silty clay loam		
Pore volume (cm^3)	8.73	8.63	8.53	8.24
Porosity (cm^3/cm^3)	0.673	0.665	0.658	0.635
Sample thickness (cm)		0.64		
Sample area (cm^2)		20.27		
Sample volume (cm^3)		12.97		

measurements were read at five or more successive 4–8 h intervals (depending on the water transfer rate) by a cathetometer device (magnitude ± 15 cm) that measured strain magnitude in the springs.

Two experimental runs were conducted with each aggregate size at each of the four temperatures. Relative humidity values were measured on each side by using psychrometer humidity indicators (accuracy$\pm 3\%$, Greenhouse Mega Store). The vapor densities were calculated by multiplying the saturation densities from a standard table by the measured relative humidities at each temperature.

4 Results and Discussion

The average rates of water vapor loss and gain for each aggregate size at each temperature are plotted in Fig. 2. These data show that the flow rate increases with increasing temperature and as the porosity increases. For the materials used in this study, the larger porosities are associated with the larger aggregates, possibly because of more irregular shapes in the larger sizes. The temperature effect is quite strong and is attributed to an increase in the kinetic energy of the water molecules, which in turn increases their velocity while diffusing and probably also reduces the length of time that they are sorbed along the way.

The water vapor losses and gains all plot as curvilinear functions of temperature and have remarkably similar curves (Fig. 2). The loss curves in particular form such a tightly knit family that one is tempted to consider them all the same. Differences in soil porosity as shown in the gain curves thus result more from differences in the amounts of water absorbed by the soil samples than from variations in the amount of water entering the samples from the humid side. The rate of water absorption at any one temperature increases with decreasing porosity. In these samples, decreasing porosity is correlated with smaller aggregate sizes and therefore with an increasing number of passageways and an increasing amount of surface area exposed to the diffusing water vapor. The lowest curve in Fig. 2 represents the same size of aggregates as its nearest neighbor represents, but with a reduction in pore space achieved by compacting the sample. This, too, should produce a larger number of smaller passageways with an increased amount of exposed surface and, probably, with an increase in the tortuosity of the passageways.

Figure 2 shows clearly that temperature has a strong effect on water diffusivity and that this effect is similar at all four of the soil porosities studied. Warmer temperatures resulted in more rapid movement of water vapor. There was an interaction between temperature and porosity on the amount of water vapor absorbed by the soil (the differences between water lost
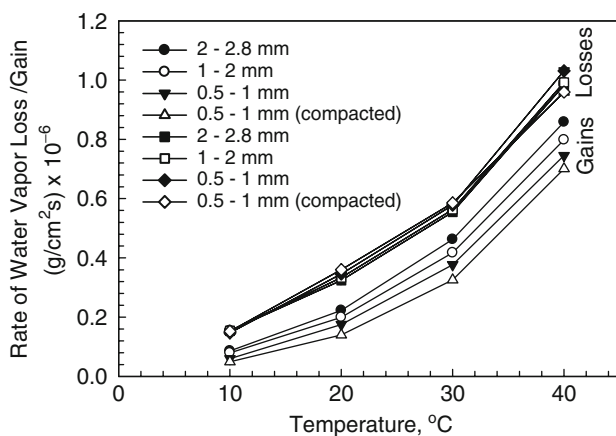
Fig. 2 The rates of water losses from the pan and water gains by the calcium chloride as influenced by temperature and size range of the Fayette silty clay loam subsoil aggregates

on the one side and water gained on the other side). The higher temperatures combined with the lower porosities resulted in much more water vapor absorption than would be calculated by adding the independent effects of the two factors.

It should be noted here that the D values for water vapor in this experiment were generally about 1/3 to 1/4 as large as those for the diffusivity of other gases that do not interact with the soil material as reported in the literature (Troeh et al. 1982). This difference can be explained by inferring that much of the diffusing water vapor was slowed by temporary sorption as it passed through the soil sample. The true diffusion coefficient should represent the water vapor while it is moving, but not while it is being held in a sorbed condition. Therefore, the diffusion coefficient determined from the data for this experiment will be designated D_a for apparent diffusion coefficient. D_a should always have a smaller value than the true diffusion coefficient, D.

The relative humidity values measured on the water side at 10, 20, 30, and 40°C were 86%, 81%, 75%, and 70%, respectively; those on the calcium chloride side were 56%, 51%, 45%, and 40%, respectively. Water vapor densities from a Handbook of Chemistry and Physics multiplied by the relative humidity differences yielded vapor pressure gradients, $C_w - C_e$, of 2.82, 5.19, 9.11, and 15.36×10^{-6} g cm^{-3} across the soil samples at the four temperatures. Table 2 shows the values of α, D_a, D_a/D_0 along with the remaining data needed for their calculation.

The values of α from Table 2 are plotted in Fig. 3. This figure shows that the rate of water absorption is a function of both temperature and size of aggregates in this soil material. The faster rate of absorption at higher temperatures shows the same trend as the absolute humidity of the air on the water side at the four temperatures (8.1, 14.0, 22.8, and 35.8×10^{-6} g cm^{-3}) but at a decreasing proportion at the higher temperatures. This decreasing proportion is attributed to faster molecular movement at higher temperatures making the water molecules more difficult to absorb and hold. The water absorption rate increases considerably as the aggregate size decreases, an effect that can be attributed in part to the increased exterior surface area resulting from the smaller size of the aggregates and in part to the greater densities of the samples composed of smaller aggregates. The higher densities have more soil and therefore more total surface area to absorb water and probably have more tortuous

Table 2 Data for the calculation of D_a and $D_a D_0$ (each value is the average of two runs)

Aggregate size	Temperature °C	$q_a \times 10^{-6}$ g cm^{-2}s^{-1}	Absorbed H$_2$O g	Hours	$\alpha \times 10^{-6}$ g cm^{-3}s^{-1}	D_a cm^{2}s^{-1}	D_a/D_0
2–2.8 mm							
	10	0.0864	0.476	96	0.1062	0.0273	0.117
	20	0.2227	0.473	64	0.1583	0.0337	0.135
	30	0.4625	0.322	48	0.1437	0.0357	0.135
	40	0.8583	0.363	40	0.1944	0.0384	0.137
1–2 mm							
	10	0.0794	0.517	96	0.1153	0.0264	0.113
	20	0.1994	0.586	60	0.2092	0.0328	0.132
	30	0.4173	0.596	56	0.2279	0.0344	0.130
	40	0.7983	0.566	40	0.3030	0.0373	0.133
0.5–1 mm							
	10	0.0600	0.611	96	0.1363	0.0235	0.100
	20	0.1750	0.697	56	0.2666	0.0321	0.129
	30	0.3769	0.821	56	0.3140	0.0335	0.127
	40	0.7452	0.835	40	0.4471	0.0370	0.132
0.5–1 mm Compacted							
	10	0.0493	0.719	96	0.1604	0.0228	0.097
	20	0.1407	0.897	56	0.3431	0.0309	0.124
	30	0.3255	0.910	48	0.4060	0.0320	0.121
	40	0.7007	0.755	40	0.4042	0.0346	0.124

Fig. 3 The effects of temperature and aggregate size on the value of α (the rate of water absorption by the soil samples)
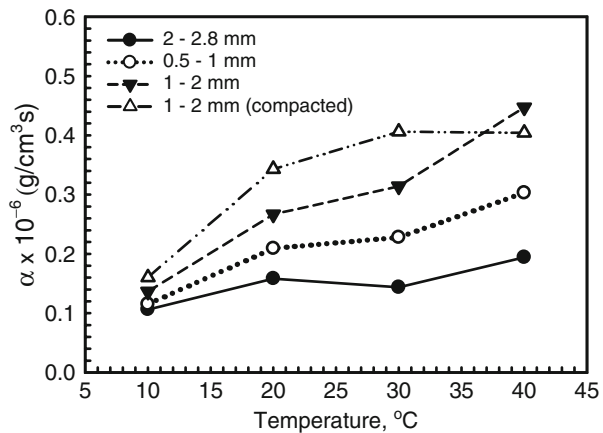
diffusion pathways resulting in slower diffusion and increased time available for absorption to occur.

The relationship between the apparent water vapor diffusivity, D_a, and soil porosity for this soil material is graphed in Fig. 4. The plotted lines for the four temperatures have similar shapes and show that both higher temperature and increased pore space contribute to faster diffusivity.

Figure 5 is a plot of D_a/D_0 versus porosity for each of the four temperatures. This dimensionless parameter removes much of the temperature effect by dividing each diffusivity value by the diffusivity in open air space at the same temperature. The resulting values for 20, 30, and 40°C are closely grouped, but the values for 10°C are considerably lower than the others.

The temperature effects are expected to follow a systematic progression from the lowest to the highest temperature, so these results must be considered anomalous. A possible explanation for the discrepancy is that the temperature control of the Percival Chamber may not have been sufficiently accurate. The 10°C temperature required cooling, and if one assumes that it might actually have been 12°C, the resulting adjustments in the $C_w - C_e$ and D_0 values would raise the 10°C line up to the approximate position of the other three lines in Fig. 5. Similarly, assuming that the 20 and 30°C temperatures were actually 18 and 29°C would place these two lines in stepwise positions between the 10 and 40°C lines. While the exact temperature is suspect, the settings appear to have been consistent from one run to the next; otherwise the data would not have plotted on lines as nearly straight and parallel as they are.

The D_a/D_0 values of the data graphed in Fig. 5 differ quantitatively from relative diffusivity values reported by various authors for non-absorbed gases (Troeh et al. 1982). For example, u and v constants to fit the data of several researchers to the diffusivity equation $D/D_0 = [(S - u)/(1 - u)^v]$ were reported by Troeh et al. (1982). The u values ranged from

Fig. 4 The effects of soil porosity and temperature on the apparent diffusivity, D_a, in aggregates from Fayette silty clay loam subsoil material
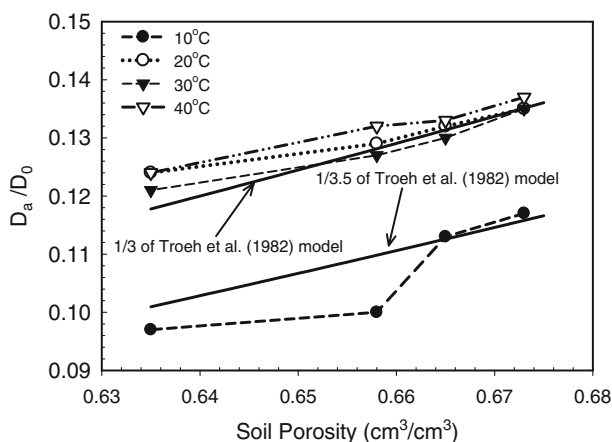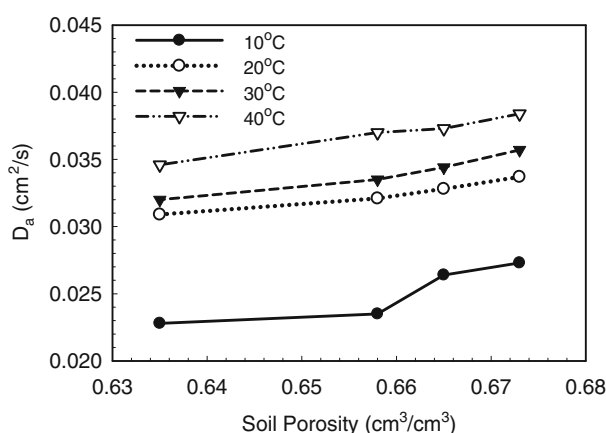

Fig. 5 The effects of soil porosity and temperature on the apparent relative diffusivity, D_a/D_0, in aggregates from Fayette silty clay loam subsoil material. The two dashed lines represent 1/3 and 1/3.5 times the diffusivity equation $D/D_0 = [(S - 0.1)/0.9]^2$

0 to 0.15 and the v values from 1.1 to 2.0. Using u and v values of 0.1 and 2.0 in this equation gives D/D_0 values from 0.353 to 0.405 for the porosity range included in this experiment ($S = 0.635$–0.673). These values are approximately three times as large as the D/D_0 values for the three closely grouped lines in Fig. 5. A probable interpretation of these data is that the diffusing water molecules actually moved at the faster rates indicated by D/D_0. The lower D/D_0 values include time delays caused by temporary sorption of the diffusing water vapor. A correction for such additional time can be made by multiplying the diffusion equation by a suitable coefficient. The two solid lines in Fig. 5 result from multiplying the values from $D/D_0 = [(S - 0.1)/0.9]^2$ by 1/3 and 1/3.5. Data points would fall on these two lines if the water molecules spent 2 or 2.5 times as long in sorbed positions as they did in actual movement. The resulting agreement with the data shown in Fig. 5 is close enough to imply that water vapor behavior in the diffusion process differs from that of other gases used in previous studies only in ways related to its being sorbed by the soil.

5 Summary and Conclusions

Water vapor diffusivity through dry soil in response to a concentration gradient increases with increasing soil porosity and rising temperature. The dry soil in this experiment absorbed from 1/8 to 2/3 of the diffusing water. Maximum absorption rates occurred with the most compact soil samples at the highest temperature, though the maximum absorption as a percentage of the diffusing water was in the compact samples at the lowest temperature. Equations derived to determine diffusion coefficients from these data include a sink term to account for the absorbed water and for the variations in the amount of water diffusing through different positions within the soil samples. Even the water that diffused all the way through the soil samples must have been slowed by temporary sorption on soil particles. It may be inferred from the data that the diffusing water molecules in this experiment spent 2 to 2.5 times as long in sorbed positions as they did in actual diffusion. The diffusivity equation $D/D_0 = [(S - 0.1)/0.9]^2$ fit the D/D_0 values obtained from these data if a coefficient of 1/3 or 1/3.5 is added to correct for the time delays caused by temporary sorption of the diffusing water vapor. The results are influenced by the interaction of water vapor with the soil materials so that the data from this study represent an apparent diffusion rate that is slower than the rate that would have occurred without such interaction. The movement of water vapor in the soil plays an important and critical role in the overall water and energy balance of the surface environment of arid and semiarid regions in many agricultural, meteorological, and engineering applications.

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